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ADSORPTION, MIGRATION, AND DISPERSION OF STRONTIUM AND CESIUM IN AN N-AREA SOIL

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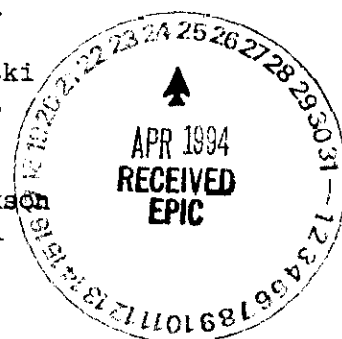
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ADSORPTION, MIGRATION, AND DISPERSION OF STRONTIUM AND CESIUM
IN AN N-AREA SOIL

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ADSORPTION, MIGRATION, AND DISPERSION OF STRONTIUM AND CESIUM IN AN N-AREA SOIL

B. F. Hajek

This report presents both experimental and mathematical results of an investigation requested by the N-Reactor Department of the Hanford Project, Richland, Washington. The research was needed to provide data for use in evaluating the potential for disposal of emergency liquid waste water to the ground as a safe and economical alternate to tank storage. The ground disposal facility being considered by the N-Reactor Department would not be used for routine disposal of liquid waste water. Use would be made only for disposal of radioactive waste coolant generated in the event of a reactor primary loop failure incident. The facility would be used for disposal of waste water from one incident.

Extensive research and development efforts in geology, hydrology, mineralogy, soil chemistry and process engineering have been applied to ground disposal problems at Hanford. Results of these studies and monitoring of large-scale disposal facilities have shown that controlled ground disposal may be conducted safely and economically in this environment.

Ion exchange relationships between soil material from the proposed site and the specific waste must be made before a final decision as to the feasibility and safety of ground disposal can be made. Therefore, objectives of this study were to determine the adsorption, elution and diffusion characteristics of trace quantities of strontium and cesium (considered to be the

limiting radionuclides) in subsurface soil material at the probable emergency disposal site and to estimate the soil percolation.

SUMMARY

Experimental and mathematically derived results are presented which characterize the adsorption, migration and dispersion in an N-Area soil of strontium and cesium present in a simulated emergency waste water.

The experimental results from laboratory soil columns and equilibrium studies showed that the soil at the proposed disposal site is more selective for cesium than strontium. The distribution coefficients were 420 and 43 ml/g for cesium and strontium respectively. Extrapolation of column breakthrough data showed that 42 and 12 column volumes of influent were required for 0.01 percent breakthrough for cesium and strontium respectively.

Calculations based on theoretical equations and equilibrium distribution coefficients indicated that the average migration rate of cesium through a soil column should be 1/10 the rate of strontium. The actual migration rate of cesium through the column was 1/11 of strontium rate. Migration rates in ground water were determined relative to ground water movement:

Migration rate of Sr = 1/100 rate of ground water movement
Migration rate of Cs = 1/1000 rate of ground water movement

Diffusion coefficients determined for strontium and cesium in soil material indicate that spreading of these adsorbed radionuclides by this mechanism is negligible.

The average infiltration rate was determined to be 170 gpd/ft^2.

MATERIALS AND METHODS

Soil Columns

The volume of emergency waste solution which can be infiltrated with-out significant strontium or cesium breakthrough was determined by the use of short laboratory soil columns through which a simulated waste liquid was perco-lated and the effluent collected for analysis. This method has been used at Hanford for several years and has been shown to give conservative field breakthrough estimates[12, 11, 8, 6]. The soil, packed into laboratory columns at field density, consisted of a composite of subsamples selected at various depths from 15 feet below the ground surface to the water table. Laboratory columns ranged from 5 to 40 cm in length. Soil material was obtained from two new test wells and from an existing well located in the vicinity proposed as the most probable location for the disposal facility. The driller's log and mechanical analysis of soil material from various depths indicated that very little variation existed between different well sites and that the subsurface material was similar to that found at the 1301-N crib[2].

The emergency aqueous waste was simulated with raw river water contain-ing traces of strontium-85 or cesium-137. Experimental soil columns were run separately with trace strontium and cesium as the uptake of a trace component by the soil usually is not affected by the presence of other trace components[4]. Strontium-85 was used as a substitute for strontium-90 since the known varia-tions in chemical and physical properties of various isotopes are slight and ion exchange reactions between isotopes can be considered indistinguishable[4]

Equilibrium Studies

Five-gram samples of the composite soil were equilibrated for 16 hours with 50 ml of the simulated waste solution or ground water traced with strontium or cesium. The suspension was centrifuged and the concentration of the radionuclide was determined in an aliquot of supernate. The difference between the equilibrium supernate concentration and the initial concentration of the radionuclide is equal to the amount adsorbed on the soil.

Diffusion

Diffusion coefficient measurements were made by filling short cylinders, closed on one end, with soil material. Soils in half these cylinders were moistened, to 20 percent by volume, with strontium or cesium traced river water. Soils in the other half were moistened to the same moisture content with untreated river water. The cylinders were joined in such a manner that soils in cylinders containing the radioactive solution were in contact with the inactive soils on the other side. After two weeks the cylinders were separated and the tracer content in each side was determined.

The mathematical solution, for ions which react with the soil, of this transient diffusion problem is[4, 10]:

$$C/C_0\,(x,\ t) = \tfrac{1}{2}\left[\mathrm{erfc}\ \frac{x}{2\left(\dfrac{D_0 t}{\theta+b}\right)^{\frac{1}{2}}}\right] \qquad [1]$$

in which -

    D_p = porous system diffusion coefficient

    θ = volumetric moisture content

    t = time

    C = activity per volume of solution

    C_o = initial activity

    x = distance from the boundary between the initially active and inactive soil

    b = adsorption factor

and

$$\text{erfc } Z = 1 - \text{erf } Z = -\frac{2}{\sqrt{\pi}} \int_0^Z e^{-\tau^2} d\tau$$

The diffusion coefficient can also be calculated from the total amount (q) of radionuclide that diffused into the initially inactive soil:

$$\frac{D_p}{\theta + b} = \left(\frac{\ell}{2}\right)^2 \frac{\pi}{t} \left(\frac{q}{q_\infty}\right)^2 \tag{2}$$

where ℓ = length of each cylinder and q_∞ = amount of material having diffused past the central plane when time is infinite. This quantity will be one-half the initial activity.

RESULTS AND DISCUSSION

Breakthrough curves for strontium and cesium are shown in Figures 1 and 2. Presentation of analytical data on logarithmic probability coördinates results in essentially a straight line which makes it possible to extrapolate the data to values below detection limits and aids interpolation between data points. Breakthrough values of less than $C/C_o = 1 \times 10^{-4}$ can be obtained in this manner.

The principal steps used to estimate disposal volumes from the resulting curves are[11]: 1) calculate the limiting C/C_o values from the ground water control limit and the concentration of radioactive strontium and cesium in the emergency waste solution; 2) locate this value on the appropriate figure and construct a horizontal line through this point, and 3) determine the waste throughput volume which corresponds to the intercept of the constructed line and the breakthrough curve. This volume is the limiting throughput volume.

Strontium will probably limit the disposal volume; however cesium could conceivably be limiting if the initial strontium activity is considerably less than cesium.

These results are applied directly to the field by determining the field column volume from the expected submerged area of the facility and the depth to ground water. It is known that this method of extrapolating results from confined laboratory columns is conservative for two reasons: 1) underground spreading that occurs will result in the actual field "column" being larger than calculated[6], and 2) log-probability breakthrough curves of long columns usually have steeper slopes than those of short columns. The latter
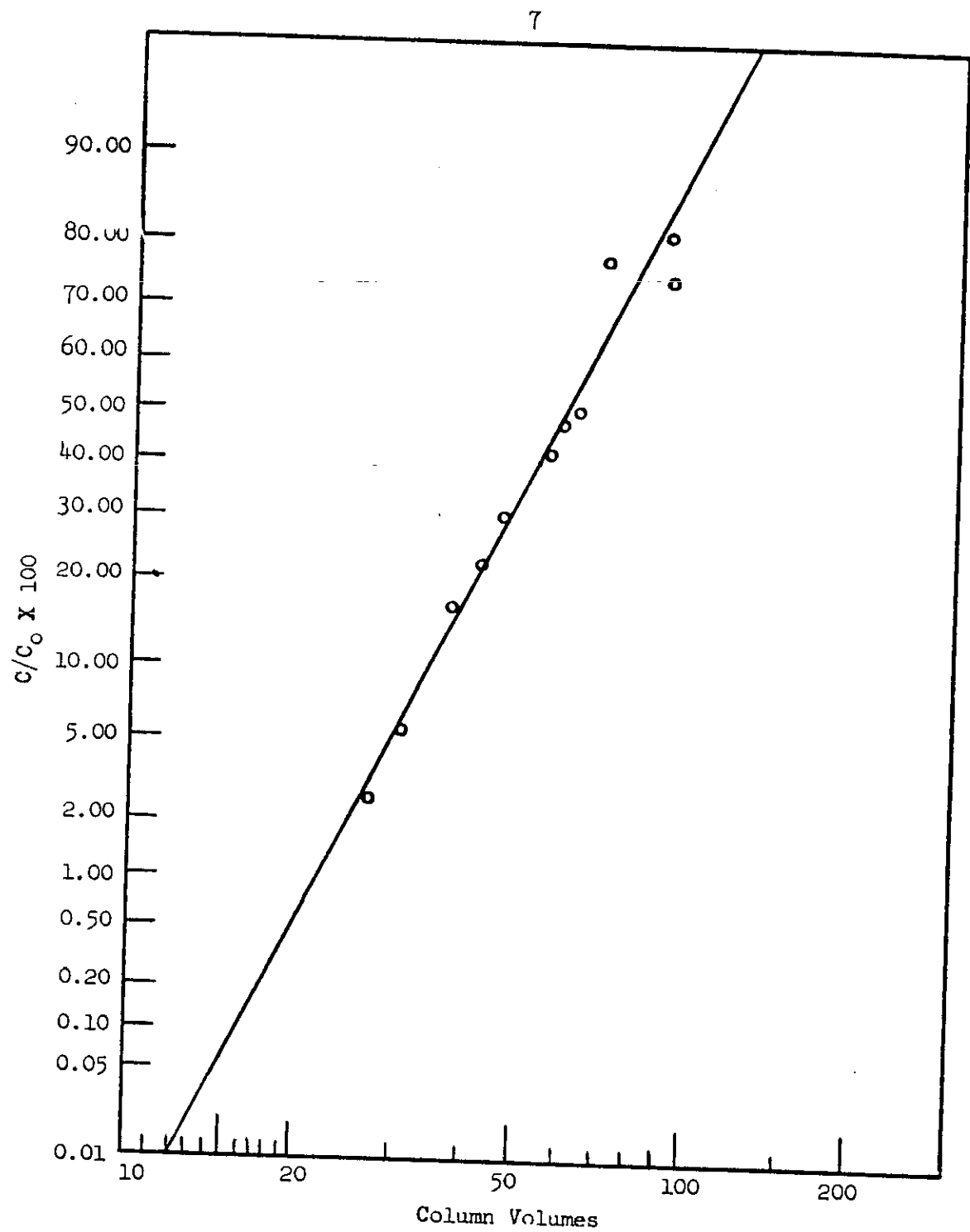
FIGURE 1

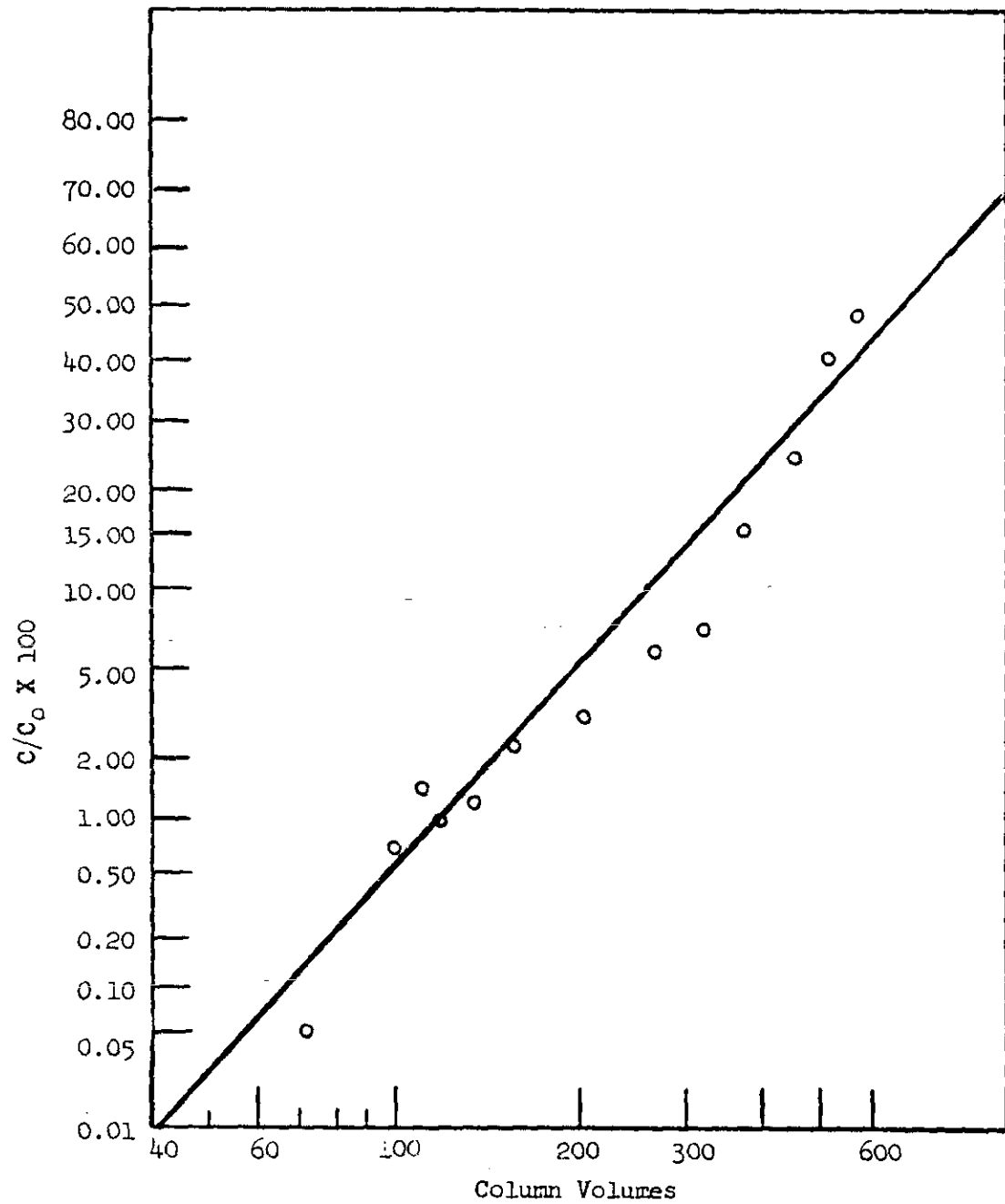Strontium breakthrough curve (Error Function C/C_o X 100 versus Log Column Volumes).

FIGURE 2

Cesium breakthrough curve (Error Function C/C_o X 100 versus Log Column Volumes).

safety factor easily could be more than two for values of $C/C_0 < 50$ percent. However, for values > 50 percent, breakthrough could occur before predicted. The lower the limiting C/C_0 value used, the greater is the safety factor.

The graphical method used to interpret these results is essentially empirical; however, the method does not include the use of any empirical constants determined with other systems. The success of this approach depends completely on the use of soil material selected from the site and a solution which adequately simulates the probable waste liquid.

Although the procedure for estimating capacity of a ground disposal facility for radioactive waste is termed "empirical," a theoretical basis does exist for breakthrough data which can be represented by a straight-line logarithmic probability curve of dimensionless breakthrough variables[13, 4, 5].

The differential equation often used to describe the concentration distribution of a solute in a one-dimensional porous media flow system is[1],

$$b \frac{\partial C}{\partial t} + v \frac{\partial C}{\partial x} = D \frac{\partial^2 C}{\partial x^2}$$

The solution of this equation with initial and boundary conditions corresponding to this study has been given as[3],

$$C/C_0 = \frac{1}{2}\left[erfc\left(\frac{x - \frac{v}{b} t}{2\sqrt{\frac{Dt}{b}}} \right) \right] + \frac{1}{2}\left[exp \frac{vx}{D} \ erf\left(\frac{x + \frac{v}{b} t}{2\sqrt{\frac{Dt}{b}}} \right) \right] \qquad [3]$$

in which:

  D = dispersion coefficient
  v = seepage velocity
  x = distance
  t = time
  b = adsorption factor

Dimensionless quantities can be introduced and the equation can be expressed as:

$$C/C_O = \tfrac{1}{2} \left[\mathrm{erfc}\left(\frac{1-\xi}{2\sqrt{\xi\varkappa}}\right) + \exp\frac{1}{\varkappa}\, \mathrm{erfc}\left(\frac{1+\xi}{2\sqrt{\xi\varkappa}}\right) \right] \qquad [4]$$

in which -

$$\xi = \frac{\bar{v}\,t}{x}$$

$$\varkappa = \frac{D}{\bar{v}\,x}$$

$$\bar{v} = \frac{v}{b} = \text{solute velocity}$$

It has been shown that for values of $\varkappa < 0.1$ the second term in the bracket is negligible[1]. In this case the equation reduces to:

$$C/C_O = \tfrac{1}{2}\, \mathrm{erfc}\left(\frac{1-\xi}{2\sqrt{\xi\varkappa}}\right) \qquad [5]$$

and a logarithmic probability plot of C/C_O versus ξ gives essentially a straight line for all values of $\varkappa < 0.1$.

Values of D may be obtained by matching experimental data to a family of C/C_O curves plotted for various values of $\varkappa$ in equation [4]. If $\varkappa$ is less than 0.1, another graphical solution is possible.

For every C/C_O obtained by experiment the corresponding value of $\dfrac{1-\xi}{2\sqrt{\xi\varkappa}}$ can be obtained from tables of error functions. Further, for every experiment ξ is known and $\dfrac{1-\xi}{2\sqrt{\xi}}$ can be calculated.

If a straight line is drawn through the points plotted on a $\dfrac{1-\xi}{2\sqrt{\xi\varkappa}}$ versus $\dfrac{1-\xi}{2\sqrt{\xi}}$ graph, the slope of the line is $\sqrt{\varkappa}$ which is equal to $\left(\dfrac{D}{vx}\right)^{\frac{1}{2}}$. Both v and x are known from column operating conditions.

When disposal into a facility terminates, it is of interest to know the concentration distribution of radionuclides in the profile bounded on top by the facility and on bottom by ground water.

Figure 3 shows a calculated soil solution concentration distribution of strontium after 6 and 12 column volumes of emergency waste have been disposed. The distribution shown was calculated for the laboratory soil column by the use of equation [5]. Accuracy of this distribution has the same limitations as listed for the breakthrough curves.

The concentration of a radionuclide in a unit volume at any point in the profile will be the sum of the nuclide adsorbed on the soil plus the equilibrium concentration in the soil solution. Downward movement of solution ions will occur even if the ionic strength of the leaching solution is very low, such as water entering the soil from precipitation. The downward movement of activity would be minor in this case, amounting to about 1/3 column volume disposal. When river water was used as an eluting solution the average downward displacement of radionuclides per column volume of eluent was the same as for a column volume of initially strontium or cesium traced river water. This type of elution is analogous to the downward movement of an activity band caused by a pulse injection. The peak activity ratio (C/C_o) and the width of the band depend on the duration of the pulse and the amount of eluent.

The elution breakthrough curves shown in Figure 4 were determined in a manner analogous to the case of a slug injection where the solute is injected for a finite time t', i.e., from zero to t_o. By introducing this time into
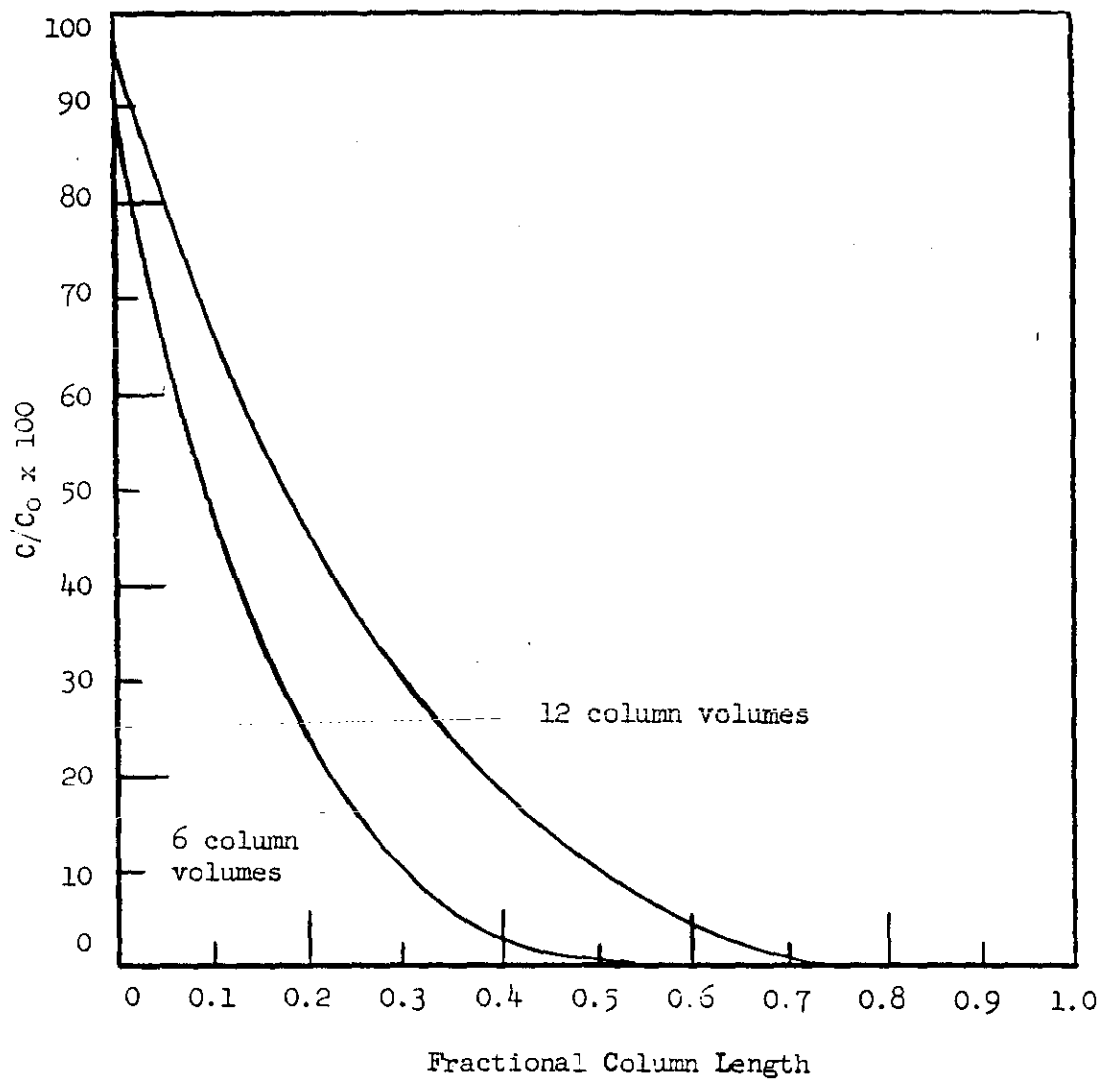
FIGURE 3

Relative Concentration of Strontium in Soil Solution after
Disposal of 6 and 12 Column Volumes of Simulated Waste.

equation [4] the equation for breakthrough of a slug injection is,

$$C/C_0 = \frac{1}{2}\left[\text{erfc}\left(\frac{1-\xi'}{2\sqrt{\xi'\varkappa}}\right) - \text{erfc}\left(\frac{1-\xi}{2\sqrt{\xi}\varkappa}\right)\right] + \frac{1}{2}\exp\frac{1}{\varkappa}\left[\text{erfc}\left(\frac{1+\xi'}{2\sqrt{\xi'\varkappa}}\right) - \text{erfc}\left(\frac{1+\xi}{2\sqrt{\xi}\varkappa}\right)\right]$$

where $\quad \xi' = \dfrac{t\,v}{b\,\varkappa}$ [6]

The equilibrium distribution coefficient (K_d) used to calculate migration rates was evaluated by the expression,

$$K_d' = \left(\frac{A\ soil}{A\ soln}\right)\left(\frac{ml\ soln}{g\ soil}\right)$$ [7]

in which;

A soil = amount of A associated with the soil

A soln = the amount of A in the equilibrium solution

ml soln = total volume of equilibrium solution

g soil = weight of soil

The equilibrium distribution coefficient is based on a mass action formulation of ion exchange. Simple stoichiometric equivalence between ions adsorbed and desorbed is generally assumed and reversibility is taken for granted. Strontium and cesium reactions in soils generally satisfy the mass action criteria and the assumptions hold, especially for the adsorption of trace quantities.

The K_d equation can be related to ion exchange theory as follows: Consider the ion exchange reaction -
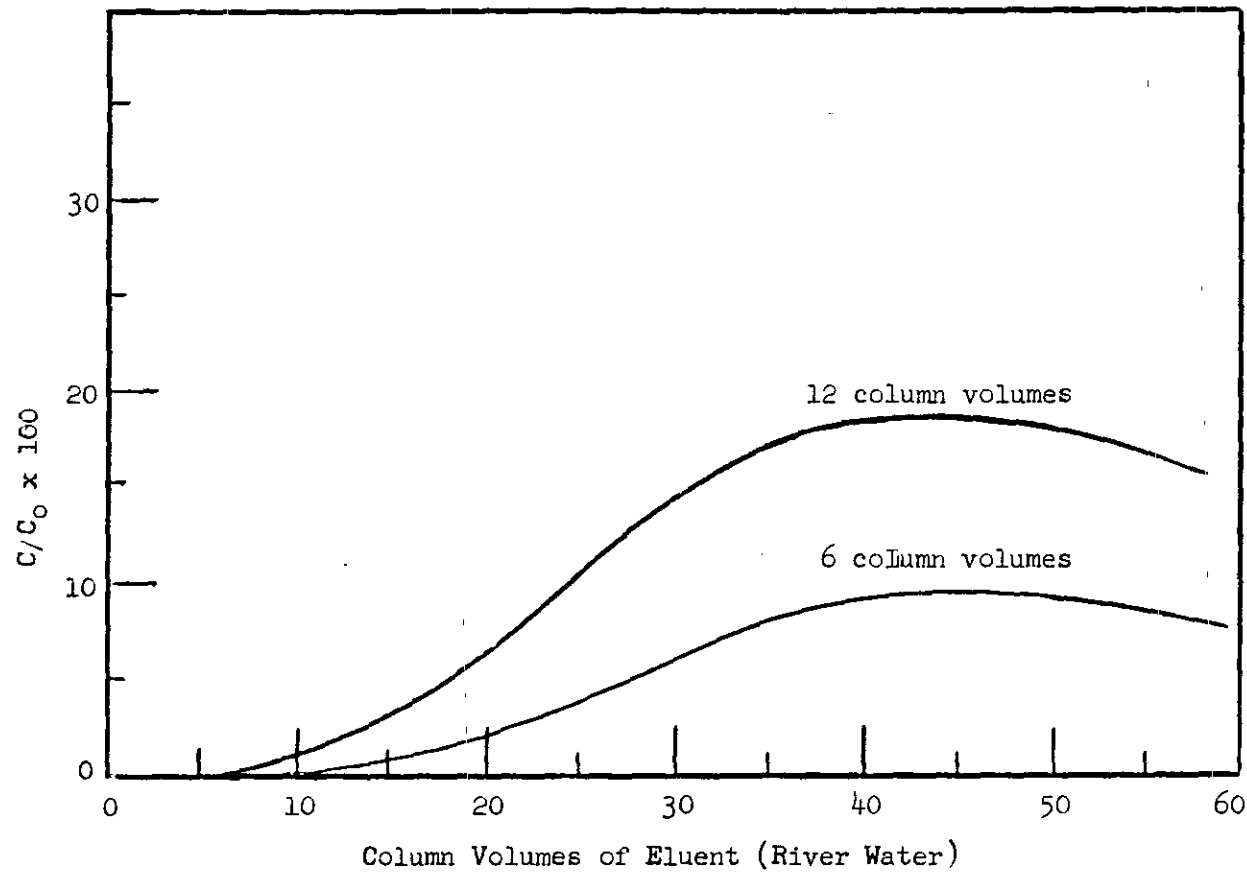
$$\text{soil Ca} + 2\text{Cs}^+ \rightleftharpoons \text{soil Cs} + \text{Ca}^{+2}$$
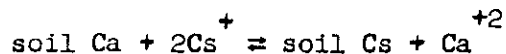
FIGURE 4

Calculated Breakthrough of Strontium Eluted by River Water After
6 and 12 Column Volumes of Waste Disposed.

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The specific mass action equation is:

$$\frac{\text{Soil Cs}}{\text{Cs}^+} = K \left(\frac{\text{Soil Ca}}{\text{Ca}^{+2}}\right)^{\frac{1}{2}} \qquad [8]$$

if soil Cs is expressed in quantity per gram and Cs^+ in quantity per ml of solution, the term,

$$\frac{\text{Soil Cs}}{\text{Cs}^+} = \text{ml}/\text{g} = K_d \qquad [9]$$

Often the quantity Soil Cs is expressed as quantity per volume (including interstitial volume) of soil in this case;

$$\frac{\text{Soil Cs}}{\text{Cs}^+} = (K_d)(\text{bulk density}) = \tau'' \qquad [10]$$

$\tau'' = $ column distribution ratio[4]. Equilibrium distribution coefficients of trace ions are easily measured and can be applied to many practical problems of radionuclide adsorption and migration in soils. The adsorption factor b in the diffusion equation is equal to τ''. At very slow flow ratios τ'' approaches b in the dispersion equation. The average migration rate of ions can be readily derived with respect to the column distribution coefficient[4, 5]. The rates reported in Table I were calculated from the expression;

$$v_i = \frac{v \text{ soln}}{1 + \frac{\tau''}{\theta}} \qquad [11]$$

where -

$$v_i = \text{migration rate of species i}$$

$$v \text{ soln} = \text{flow velocity through pores}$$

The ratio of distribution coefficients indicates the extent of separation

TABLE I

Summary of Adsorption, Migration and Diffusion Characteristics

Diffusion Coefficients (D_p)

$$\text{strontium} \qquad 1.4 \times 10^{-7} \text{cm}^2/\text{sec}$$

$$\text{cesium} \qquad 2.1 \times 10^{-7} \text{cm}^2/\text{sec}$$

Adsorption Factors (b)

$$\text{strontium} \qquad 73$$

$$\text{cesium} \qquad 712$$

Migration rate relative to flow of soil solution

$$v_{\text{strontium}} = \frac{v_{\text{solution}}}{100}$$

$$v_{\text{cesium}} = \frac{v_{\text{solution}}}{1000}$$

Effect of NH_4^+ on migration rate

$$v_{\text{strontium}} = \frac{v_{\text{solution}}}{100}$$

$$v_{\text{cesium}} = \frac{v_{\text{solution}}}{800}$$

___

of two radionuclide species as they migrate through a soil column[4]. This relationship is easily seen in the column and equilibrium experimental results of this study. The distribution coefficients for adsorption, by the composite soil, of cesium and strontium from river water were 420 and 43, which equals a separation of 10. The measured column separation (migration rate of the 50 percent point) was 11. The distribution coefficients of cesium and strontium in ground water and soil material from the ground water strata were 189 and 19

respectively, a separation of 10 which agrees well with the other determinations.

During routine N Reactor operations, a liquid waste containing 10 ppm NH_4^+ is disposed to the ground in the 1301-N waste disposal crib. None of this waste will reach sediments beneath the proposed disposal facility. However, the ammonium ion enters ground water beneath the crib at a point between the planned site and the Columbia River. If strontium or cesium should enter the ground water at the site being studied, eventually these ions would migrate into sediments which nave been contacted by ammonium containing waste. The ammonium ion is known to compete favorably for cesium exchange sites and could affect the migration rate of cesium significantly.

A study was conducted in which 10 ppm NH_4^+ was added to ground water traced with cesium and strontium which then was equilibrated with soil material from the ground-water strata. The data showed (Table I) that the ammonium ion at this concentration had no effect on trace strontium adsorption; however, the cesium distribution coefficient was reduced to 153 and consequently the separation factor is reduced and the velocity of cesium in this system would increase. (Table I).

The diffusion coefficients measured indicate that the degree of strontium and cesium spreading by diffusion would be so small that this mechanism of spreading can be ignored. The only possible incident of ground water contamination by diffusive movement of radionuclides would be if the adsorbed zone were very near the water table. However, if this occurred the activity released to ground water would be minor due to the slow rate of movement.

It is not possible to calculate an average migration rate due to diffusive movement. This is a dispersive action and thus each concentration plane in the soil profile will migrate at a different rate which will not be constant.

An estimate can be made by assuming an initial distribution in the column profile as shown in Figure 3 and the use of equation [1].

The diffusion coefficients (D_p) and adsorption factors (b) used are shown in Table I. The volumetric moisture content (θ) was 20 percent. The variable t (time) in equation [1] can be replaced by $t_o + t$, in which t_o is the time it would require to achieve the initial distribution by diffusion. If 6 column volumes were disposed, the 5 percent concentration plane would migrate about 1/100 of the column length in the first 1000 years, following disposal, or about 0.5 feet in a column 50 feet long. Diffusion of cesium would be less.

The permeabilities calculated from the driller's infiltration data are shown in Table II. These data, expressed in terms of gallons/day/square foot under unit head and a hydraulic gradient of 1.0, are an approximation of saturated permeability. The intake tests were conducted under unsaturated conditions and are not valid saturated permeability values. The soil column at the proposed facility is not expected to become saturated before disposal terminates Thus an intake value other than the saturated permeability of the least permeable strata is needed for design. The infiltration value shown in Table II is suggested as the design intake value for disposal of a limited amount of waste. This value is based on both permeability and other data[14] obtained from actual waste disposal facility performance in the 100 and 200 Areas of the Hanford Project.

TABLE II

Some Disposal Characteristics of N-Area Soils

Permeability*

Depth Ft	Permeability gpd/ft^2
17	108
28	27
39	461

*Based on analysis of data by a method developed by the Bureau of Reclamation (Engineering Monograph #8)

Infiltration

Approximate infiltration rate based on test well and other data

170 gpd/ft^2

Depth to Ground Water

Well No.	Casing Elev.	Ground Water
N-6	460.405	402.625
N-7	460.160	403.480
86-60	453.07	404.87

Grain Size Analysis

Particle size	Weight %
> 2 mm	40
2 - 0.05 mm	43
0.05 - 0.002 mm	15
< 0.002 mm	2

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